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Permeability and Compression Characteristics of Clay Contaminated with Kerosene and Gasoil

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Abstract

Pore fluid chemistry plays an important role in physical and mechanical behaviour of clayey soils. In the present study, physical and index tests were performed along with series of one-dimensional consolidation tests to evaluate the effect of two organic fluids including kerosene and gas oil as contaminant, on the behaviour of clayey soil. Kaolinite and montmorillonite were used as the base soils in the tests. Three different weight percentages of contaminants (3%, 6% and 9%) were used in sample preparation. The samples were prepared with two relative compactions of 70% and 90% and one-dimensional consolidation tests performed to investigate the compressibility behaviour of the contaminated soils. Results show the increase in compressibility of contaminated clay due to the change in chemical characteristics of pore fluid. Also as a consequence of higher chemical activity, the diffuse double layer thickness increases in contaminated clays, while the consolidation rate parameters including consolidation coefficient and permeability decrease with higher contaminant percentage.

1. Introduction

Soil contamination is not only important as an environmental issue, but it is also considerable from geotechnical perspective due to the consequences which can alter the soil properties. Contamination in soils arises from variety of sources including rupture of underground storage tanks, application of pesticides and percolation of contaminated surface water to subsurface strata, oil and fuel dumping, leaching of wastes from landfills or direct discharge of industrial wastes into the soil. The most common chemicals involved in soil contamination are petroleum hydrocarbons, solvents, pesticides, lead and other heavy metals. The occurrence of this phenomenon is correlated with the degree of industrialization and intensities of chemical usage. In USA there are approximately 555,000 underground storage tanks (USTs) which store petroleum or hazardous substances (USEPA 2017). There is always potential of leakage from the storage tanks to the soil and underground water.

In 2010, the explosion of a crude oil well in the Gulf of Mexico, which was one of the worst environmental disasters in American history (EPA, 2017), led to soil pollution in the coasts of the Gulf. It was one of the obvious examples of soil contamination by the organic fluids. According to the estimations the explosion discharged about 210 million gallons of oil into the Gulf of Mexico over a period of approximately three months [1].

Physical characteristics of soils change with introduction of chemical contaminants. In such contaminated soils, this can affect slope stability, bearing capacity of foundations and other structures built on the soil. There are studies which have been carried out about contaminated soils by organic fluids including hydrocarbon liquids [2, 3] have investigated the geotechnical parameters of contaminated granular soils. A number of other researchers have investigated the mechanical behavior of fine grained contaminated soils. Fernandez and [4-8], among others investigated the effect of organic fluids on the hydraulic conductivity of clays and permeation of compacted clay with organic chemicals.[9-14] studied compressibility behavior of soils contaminated with organic fluids studied triaxial behavior of Kaolinite clays

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with different pore fluids [15]. A number of other researchers including [16-22] investigated the physical and mechanical behavior of fine-grained contaminated soils.

Study of the compressibility and other physical characteristics of clayey soils shows differences in experimental results. [18] reported the increase of plasticity index of soil by addition of organic fluids while [17] investigated the geotechnical properties of clayey and sandy soils contaminated with oil and reported a reduction in permeability, strength, maximum dry density, optimum water content, and Atterberg limits of these soils. They also found that the organic fluids caused the clay particles to flocculate and consequently it decreased settlement time of suspended particles in sedimentation tests. They correlated the low dielectric constant of toluene and ethylene glycol with flocculation of clay particles. They concluded that increase in organic fluid/water ratio or, on the other hand, decrease in dielectric constant of the pore fluid decreases the liquid limit and consolidation coefficient of the contaminated soil while it increases the shear strength value.

Generally, different studies have shown that clays are very sensitive to pore fluid chemical and the ionic concentration. The properties of pore fluid can affect the structure of the clay and the mineral-mineral contacts [23] and eventually it can affect the physical and mechanical behavior of the clay. Soil particles coated with activated sludge seem to develop a kind of hydrophobic character.

This research has focused on clay contamination with kerosene and gasoil (also known as diesel fuel) which are widely used in Iran as fuel. The knowledge about compressibility behavior of clays contaminated with kerosene and gasoil is very little. The contaminants are applied to two different types of clayey soils with different mineralogy including Kaolinite and Montmorillonite as representatives of low plasticity and high plasticity soils, respectively. The contaminants were mixed with oven dried soil in three weight percentages of 3%, 6% and 9% and the samples prepared in two relative densities of 70% and 90%. One dimensional odometer tests [14] were used to investigate the compressibility and permeability characteristics of the contaminated clays with different contaminant contents.

2. Test methods and materials

2.1. Materials

Two types of clays including montmorillonite and kaolinite, which are representative of soils with high and low plasticity, were used as base materials in the experiments. Physical properties of kaolinite and montmorillonite are presented in table 1. According to the unified soil classification system (USCS), these two soils are classified as CH and CL, respectively. The Kaolinite obtained from Hesarak area in Karaj, Iran and the Montmorillonite was obtained from Shahr-e- Rey, south of Tehran, Iran.

Table 1. Specification of clays used in the tests

Base Soil	Soil Name	LL (%)	PL (%)	PI (%)	G_s	$\gamma_{d,\max}$ kN/m^3	$w_{opt.}$ (%)
Kaolinite	CL	32	22	10	2.66	16.5	13
Montmorillonite	CH	238	66	172	2.02	11.7	33

Kerosene and Gasoil which are widely used in the vehicles, industry and heating systems in Iran, were used as two main contaminants. They are among the most probable soil contaminants in areas such as gas stations and fuel reservoirs in factories and residential complexes. Because of existence of old reservoirs in Iran, leakage of these fuels to the soils under the reservoirs is very common issue.

Kerosene is a combustible hydrocarbon liquid which is derived from petroleum, widely used as a fuel in industry as well as households. It is a thin, clear liquid formed from hydrocarbons obtained from the fractional distillation of petroleum between 150°C and 275°C, resulting in a mixture with a density of 0.78–0.81 gr/cm^3 composed of carbon chains that typically contain between 6 and 16 carbon atoms per molecule. It is miscible in petroleum solvents but immiscible in water. Gasoil is produced from the fractional distillation of crude oil between 200°C and 350°C at atmospheric pressure, resulting in a mixture of carbon chains that typically contain between 8 and 21 carbon atoms per molecule [24]. It is composed of about 75% saturated hydrocarbons primarily paraffins including n, iso, and cycloparaffins, and 25% aromatic hydrocarbons including naphthalene and alkyl benzenes [25]. The average chemical formula for common diesel fuel is $C_{12}H_{24}$, ranging approximately from $C_{10}H_{20}$ to $C_{15}H_{28}$.

Both of the contaminants are commercially available in Iran. Gasoil (diesel) is the main fuel for the trucks in Iran and it is also widely used along with kerosene in heating systems in rural areas where there is not access to natural gas.

Regardless of the crude oil source or processing history, kerosene's major components are branched and straight chain alkanes and naphthenes (cycloalkanes), which normally account for at least 70% of the kerosene's volume. Aromatic hydrocarbons in this boiling range, such as alkylbenzenes (single ring) and alkylnaphthalenes (double ring), do not normally exceed 25% by volume of kerosene streams. Olefins are usually not present at more than 5% by volume. Although its chemical characteristics can be somewhat variable in different parts of the world, it has been as the contaminant in a number of researches on contaminated soils. Properties of kerosene and gas oil are shown in table 2.

Table 2. Specification of liquids used in the test

Liquid	Dielectric Constant	Viscosity (centistokes)
Water	80	1
Kerosene	1.8	2.71
Gasoil	2	13.91

2.2. Sample preparation

In order to prepare samples, the kaolinite and montmorillonite were passed through a #200 with the equivalent diameter of 0.075 mm and then the soils were placed at the oven with 110° C temperature for 16 hours to dry. The oven dried soil then was mixed with the contaminants in three different weight percentages of 3%, 6% and 9%. Also, some samples were prepared using 100% contamination. Mixing the soil and contaminant were continued to achieve a homogenized mixture of contaminated soil. To avoid evaporation of contaminant, the mixture was kept in a closed container and was wrapped with the polyethylene sheet. The mixture was kept for 7 days to reach chemical stability. Samples of soils were prepared in two different relative compactions of 70% and 90%. Relative compaction of specimens was calculated according to Eq. (1) as proportion of specific gravity of prepared specimen (G_s) to maximum dry density (γ_d) which was derived from standard Proctor tests.

$$R_c = \frac{\gamma_d}{\gamma_{d,\max}} \tag{1}$$

2.3. Test procedure

The experiments have performed in two main categories including physical and compression tests. Physical tests include liquid limit, plastic limit, specific gravity and standard Proctor tests.

In order to perform one-dimensional consolidation experiments, after preparation of soil mixture, the required weight of mixture was calculated according to the considered relative compaction and volume of the mold.

The molds containing specimens were placed in a chamber of distilled water to let them saturate for 96 hours by normal saturation method. A number of tests were performed on samples with 100% contamination at which contaminant was used to saturate specimens. Loading was started after device calibration in increments of 50 kN/m^2. The tests were performed in accordance with ASTM D2435 method.

3. Results of the tests

3.1. Physical test results

Figures 1 and 2 show the variations of Atterberg limits of contaminated kaolinite and montmorillonite. These figures show increases in liquid limit and plastic limit of contaminated montmorillonite with increase in contaminant content. This increase is not considerable in kaolinite. The increase in liquid limit can be attributed to the higher viscosity of kerosene and gas oil when compared with water as depicted in table 2. Higher viscosity of pore fluid may result in more adhesion between soil particles.

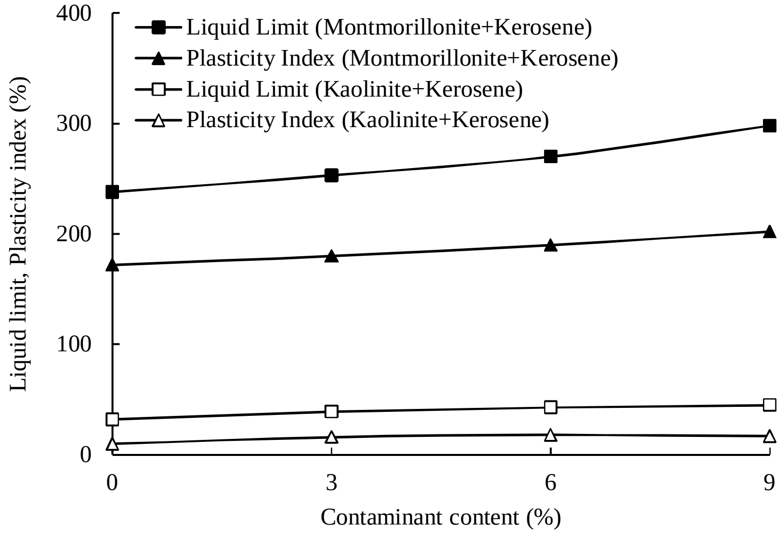

Figure 1. Liquid limit and plasticity index of montmorillonite and kaolinite contaminated with kerosene

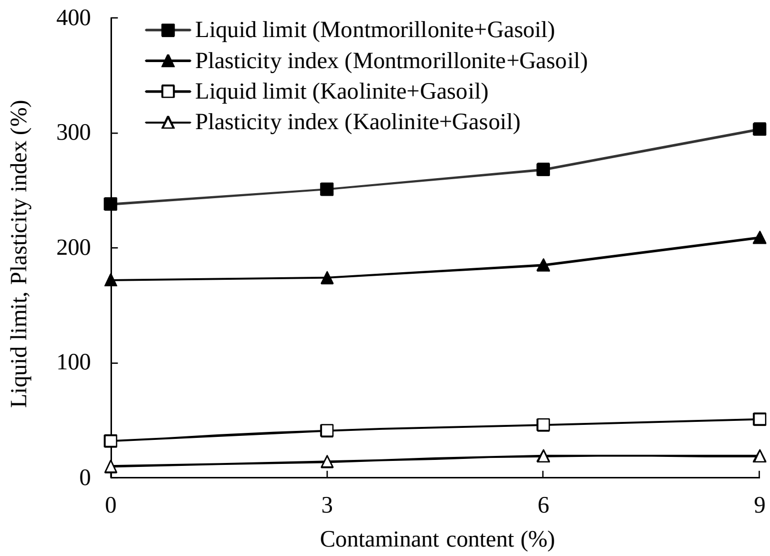

Figure 2. Liquid limit and plasticity index of montmorillonite and kaolinite contaminated with gasoil

As stated in a number of previous studies, the lower dielectric constant of contaminant decreases the thickness of diffuse double layer (DDL) and results in a flocculated structure and reduction of the Atterberg limits. However, for kerosene or gasoil contaminations, viscosity effects are stronger and the resultant effect is the increase in Atterberg limits [26].

Figure 3 represents the results of standard Proctor tests for contaminated montmorillonite and kaolinite. As shown in figure 3, the maximum dry density of montmorillonite and kaolinite are 11.7 and 16.5 kN/m^3, respectively.

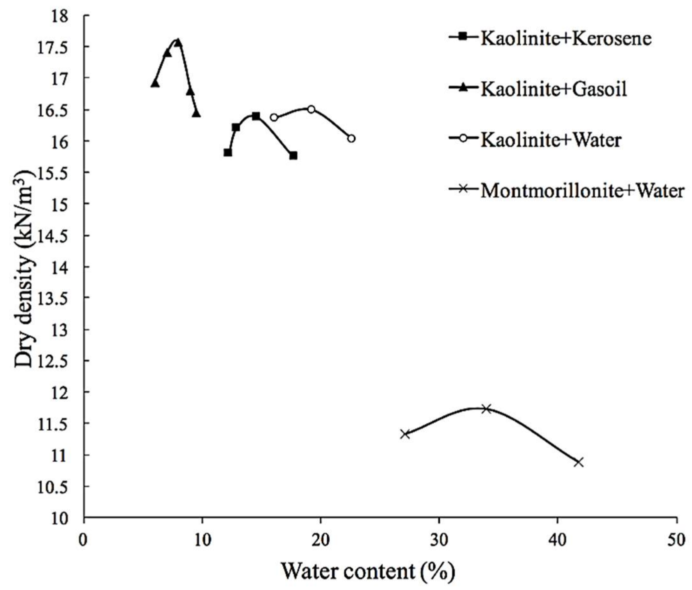

Figure 3. Results of standard Proctor tests on kaolinite and montmorillonite including water, kerosene and gasoil as pore fluid

According to the test results, compaction curves moved toward lower optimum water contents due to the contamination while maximum dry density did not change considerably for kerosene contamination. On the other hand, in the case of contamination with gasoil, other than intensive loss of optimum water content, maximum dry density increased considerably which shows more compaction due to the lubrication of clay particles. Lubrication facilitates movement of clay particles and therefore soil can be compacted more. Regarding more viscosity of gasoil compared to kerosene, decrease in optimum moisture is more when contaminant fluid is gasoil. Consequently, the specimen became more compacted in less optimum water content.

[18] Reported decrease in optimum water content and increase in maximum dry density in the case of contamination with crude oil which is consistent with results of present research. Nevertheless, [17] added different percentages of crude oil to a low plasticity clay and reported decrease in both optimum moisture and maximum dry density. The decrease of maximum dry density in [17] experiments can be related to the higher contaminant percentages compared to [18] and present research.

3.2. One dimensional consolidation tests

One-dimensional consolidation experiments were performed on kaolinite samples with relative compactions of 70% and 90% and four contaminant contents of 0%, 3%, 6% and 9%. In all tests after preparing the samples, saturation was performed using distilled water for at least 96 hours to reach 100% saturation. Figures 4 and 5 present changes in

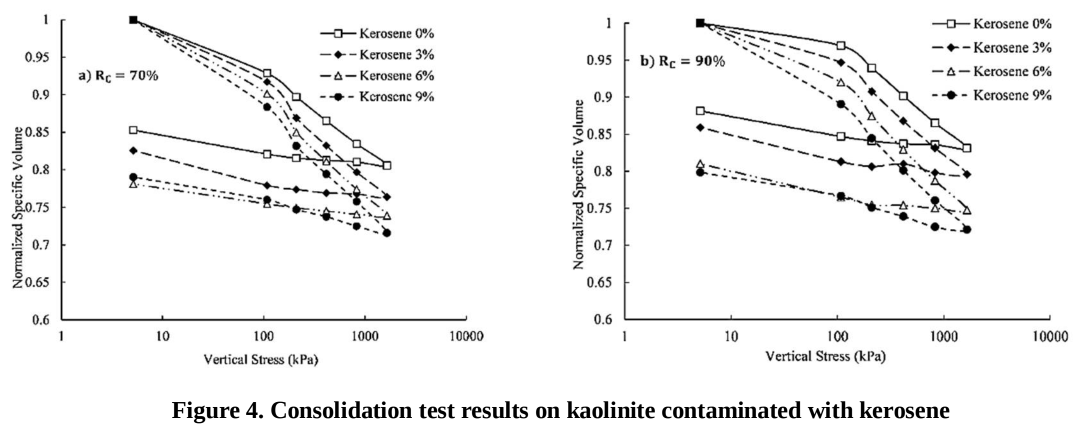

Figure 4. Consolidation test results on kaolinite contaminated with kerosene

(a) $R_c = 70\%$ **(b)** $R_c = 90\%$

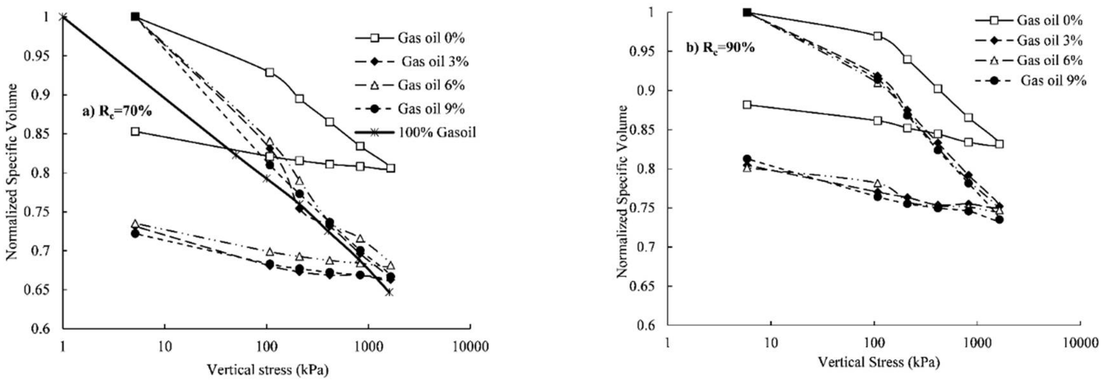

Figure 5. Consolidation test results on kaolinite contaminated with gasoil

(a) $R_c = 70\%$ **(b)** $R_c = 90\%$

Normalized specific volume (v=1+e) with variation of applied vertical stress for kaolinite contaminated with kerosene and gasoil, respectively at different relative compactions. The specific volume values have been normalized to the initial specific volume. Also, some tests included unloading-reloading segments. Based on the figures, it can be concluded that at a specific vertical stress during the consolidation tests, the specific volume decreased with increase in contaminant content. It is due to higher viscosity of gasoil and kerosene compared to the distilled water which leads to lubrication and reduction of friction between soil particles and subsequent reduction of volume under applied surcharge pressures. Consequently, compression of clay particles was being facilitated and specific volume decreased more in comparison to pure water as pore fluid. Figure 6 shows variations of compression coefficient of kaolinite contaminated with kerosene and gasoil at different relative compactions. The compression coefficient (C_c) is defined based on the following equation:

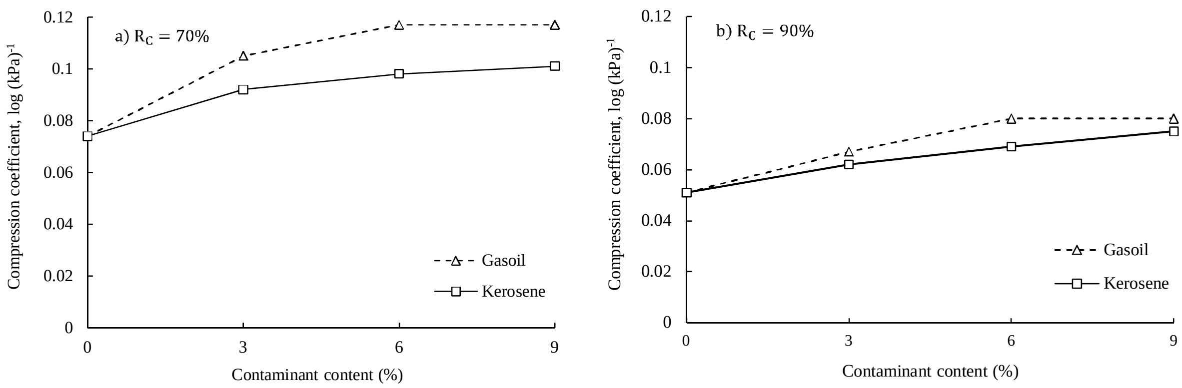

Figure 6. Variation of compression coefficient of kaolinite contaminated with kerosene and gasoil

(a) $R_c = 70\%$ **(b)** $R_c = 90\%$

$$C_c = \frac{\Delta e}{\Delta \log \sigma_v} \tag{2}$$

Based on the results, it was observed that compression coefficient of kaolinite at the relative compaction of 70% and gasoil contamination percentage of 9% increased 58% compared to the clean soil. This value was dropped to 36% for 9% kerosene contamination. According to the figures, the effect of contaminant type on compaction coefficient was less in higher relative compaction values. Additionally, the difference between compression coefficients of kaolinite contaminated with kerosene and gasoil decreased with increase in relative compaction from 70% to 90%. It is consistent with the results of [18].

The coefficient of consolidation (C_v) was calculated based on the method proposed by [27] and the results are presented in figures 7 and 8 for kerosene and gasoil contaminations, respectively. The coefficient of consolidation can be defined based on the following equation:

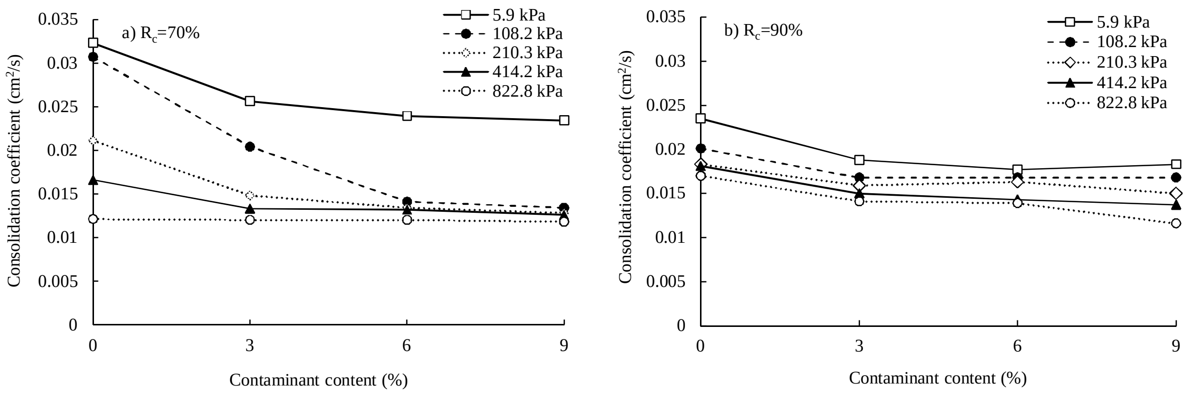

Figure 7. Variation of consolidation coefficient for kaolinite contaminated with kerosene

(a) $R_c = 70\%$ **(b)** $R_c = 90\%$

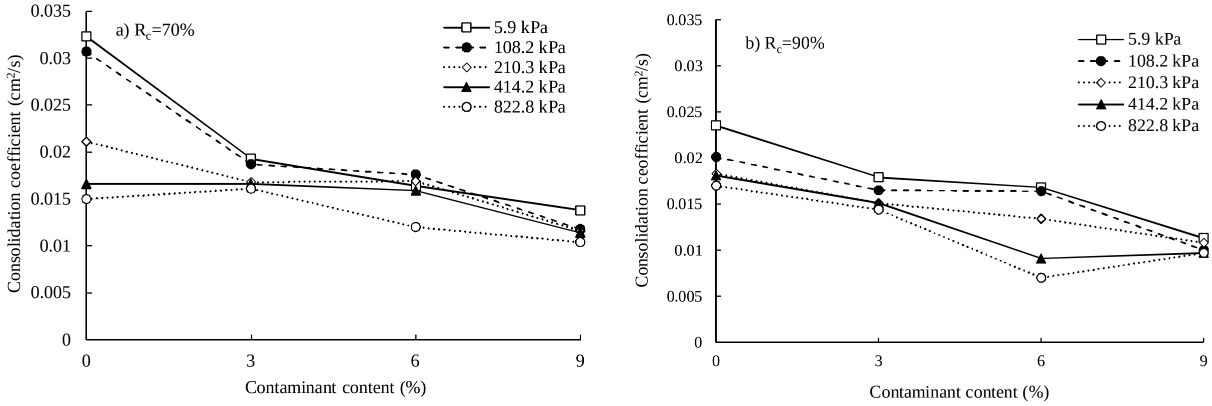

Figure 8. Variation of consolidation coefficient for kaolinite contaminated with gasoil

(a) $R_c = 70\%$ **(b)** $R_c = 90\%$

$$C_v = \frac{H^2 T_v}{t} \tag{3}$$

As it is shown in figures 7 and 8, coefficient of consolidation decreased with increase of surcharge stress. Also, increase in contaminant content resulted in decrease of consolidation coefficient. For both kerosene and gasoil contaminated kaolinite, samples with lower relative compaction had higher consolidation coefficients.

Figure 9 shows variations of the mean consolidation coefficients obtained in different surcharge pressures with contaminant content. As it is shown, the values of consolidation coefficient were higher for kaolinite contaminated with kerosene compared to the gasoil. It can be attributed to the lower viscosity of kerosene compared to gasoil which results in slower flow of gasoil within the voids and lower rate of consolidation.

The permeability coefficient (k) of contaminated kaolinite was determined based on consolidation coefficients and equation (4):

$$k = C_v m_v \gamma_w \tag{4}$$

Figures 10 and 11 summarize the permeability coefficient of kaolinite contaminated with kerosene and gasoil at different relative compactions. It can be concluded that the permeability of kaolinite decreases slightly when it is subjected to the contaminants. The decrease in permeability coefficient was more in lower surcharge pressures. In relative compaction of 70%, the permeability coefficient of kaolinite contaminated with kerosene was more than those with gasoil. The reason for this phenomenon is higher viscosity of gasoil which makes the water flow more difficult through the interconnected voids within the soil. In relative compaction of 90%, there is no significant difference between permeability coefficients of kaolinite contaminated with kerosene and gasoil which suggests minor effects of contamination on compressibility behavior of clay in higher relative densities. Indeed, at higher relative densities, the void ratio of kaolinite is very low and it leads to lower flow rate of viscous contaminant pore fluid through the interconnected voids.

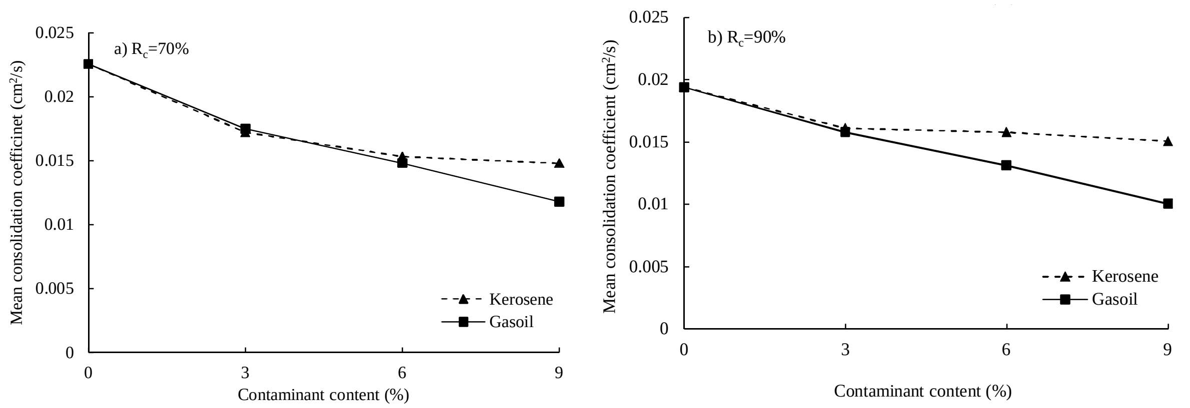

Figure 9. Variations of the mean consolidation coefficients obtained in different surcharge pressures with contaminant content.

(a) $R_c = 70\%$ **(b)** $R_c = 90\%$

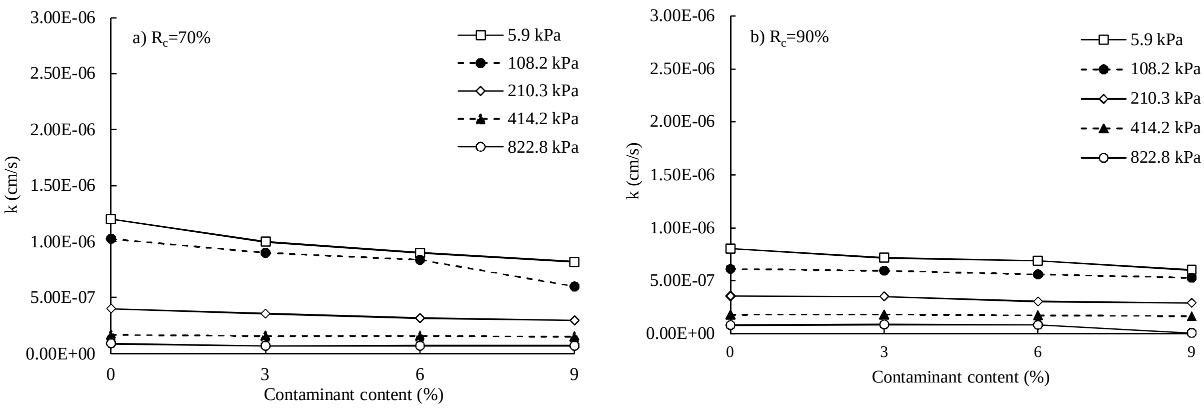

Figure 10. Variation of permeability coefficient for kaolinite contaminated with kerosene

(a) $R_c = 70\%$ **(b)** $R_c = 90\%$

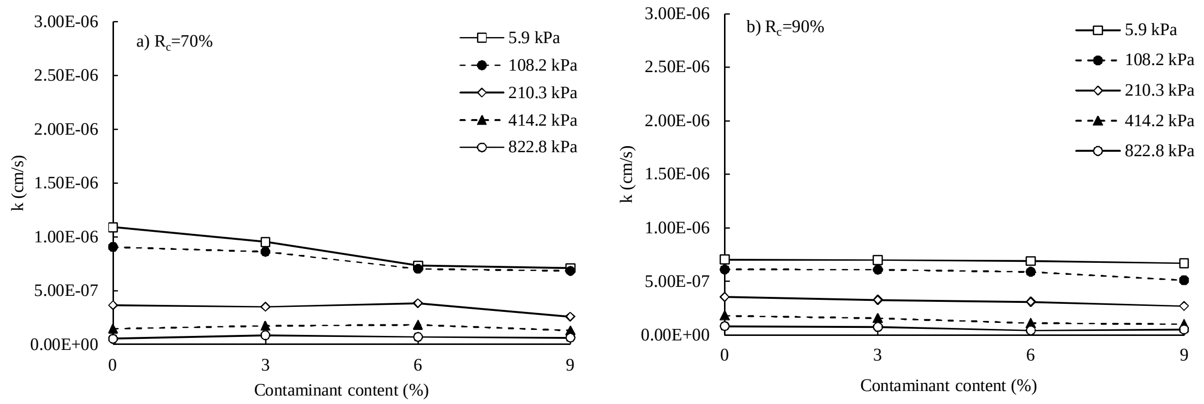

Figure 11. Variation of permeability coefficient for kaolinite contaminated with gasoil

(a) $R_c = 70\%$ **(b)** $R_c = 90\%$

4. Conclusions

A series of consolidation tests were performed on kaolinite clay contaminated with kerosene and gasoil along with physical tests on contaminated kaolinite and montmorillonite and the following results obtained. It should be emphasized that the conclusions are valid for the considered soils and applied contaminant types.

- The contamination decreased optimum water content and increased the maximum dry density of kaolinite and montmorillonite.

- The liquid limit of both considered clays increased with contaminant addition. Gasoil increased liquid limit more than kerosene.
- During one dimensional consolidation tests at a particular vertical stress level, there was a reduction of specific volume in contaminated and clean kaolinite. Also, increase in contaminant content tended the consolidation curves to lower values of specific volume.
- The compression coefficient (Cc) value increased with increase in contaminant content and was higher for gasoil contamination compared to kerosene which shows more susceptibility to the settlement. However, increase in relative compaction decreased the effect of contaminant type on compression index value.
- The consolidation coefficient (Cv) of kaolinite decreased with increase of contaminant content. The value was less when the contaminant was gasoil. For the samples with higher relative compaction, it was lower and the rate of consolidation was less consequently.
- The permeability of clays slightly decreased with increase of contamination. The reduction was more considerable in lower surcharge pressures.

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